

#16  
10/10/03  
BSDECLARATION

I, Ryuichi YAMADA, a Japanese Patent Attorney registered No. 7898 having my Business Office at Hasegawa Bldg., 4F, 7-7 Toranomom 3-chome, Minato-ku, Tokyo, Japan, solemnly and sincerely declare:

That I have a thorough knowledge of Japanese and English languages; and

That the attached pages contain a correct translation into English of the specification of the following Japanese Application:

APPLICATION  
NUMBER

DATE OF  
APPLICATION

292490/2000(Pat.) 26/SEP/2000

Applicant(s)

CANON KABUSHIKI KAISHA

Signed this ~~16<sup>th</sup>~~ day of September, 200<sup>3</sup>.

  
Ryuichi YAMADA

PATENT OFFICE  
JAPANESE GOVERNMENT

This is to certify that the annexed is a true  
copy of the following application as filed with this Office.

<u>APPLICATION NUMBER</u>	<u>DATE OF APPLICATION</u>
292490/2000(Pat.)	26/SEP/2000

Applicant(s)

CANON KABUSHIKI KAISHA

19/OCT/2001

Director-General,  
Patent Office : KOZO OIKAWA (Seal)

Certificate No. 3091959/2001

&lt;FILE NO.&gt; 4315006

&lt;DOCUMENT&gt; PATENT APPLICATION

&lt;FILE NO.&gt; 4315006

&lt;FILING DATE&gt; 26/SEP/2000

&lt;DIRECTED TO&gt; The Director General of the Patent Office

&lt;INTERNATIONAL CLASSIFICATION&gt; H05B 33/00

&lt;TITLE OF THE INVENTION&gt; LUMINESCENCE DEVICE

&lt;NUMBER OF CLAIMS&gt; 2

&lt;INVENTOR(S)&gt;

<Address> c/o CANON KABUSHIKI KAISHA  
3-30-2 Shimomaruko, Ohta-ku, Tokyo

&lt;Name&gt; Akira TSUBOYAMA

&lt;Address&gt; Ditto

&lt;Name&gt; Shinjiro OKADA

&lt;Address&gt; Ditto

&lt;Name&gt; Takao TAKIGUCHI

&lt;Address&gt; Ditto

&lt;Name&gt; Takashi MORIYAMA

&lt;Address&gt; Ditto

&lt;Name&gt; Jun KAMATANI

&lt;Address&gt; Ditto

&lt;Name&gt; Hidemasa MIZUTANI

&lt;APPLICANT&gt;

&lt;IDENTIFICATION NO.&gt; 000001007

&lt;NAME&gt; CANON KABUSHIKI KAISHA

&lt;ATTORNEY&gt;

&lt;IDENTIFICATION NO.&gt; 100096828

&lt;PATENT ATTORNEY&gt;

&lt;NAME&gt; Keisuke WATANABE

&lt;TEL&gt; 03-3501-2138

&lt;DESIGNATED ATTORNEY&gt;

&lt;IDENTIFICATION NO.&gt; 100059410

&lt;PATENT ATTORNEY&gt;

&lt;NAME&gt; Yoshio TOYODA

&lt;TEL&gt; 03-3501-2138

&lt;OFFICIAL FEES&gt;

&lt;PREPAY REGISTRATION NO.&gt; 004938

&lt;AMOUNT&gt; 21,000

&lt;ARTICLES PRESENTED&gt;

&lt;ARTICLE&gt; Specification 1

&lt;ARTICLE&gt; Drawing(s) 1

&lt;ARTICLE&gt; Abstract 1

&lt;GENERAL POWER NO.&gt; 9703710

&lt;REQUEST FOR PROOF&gt; YES

[Document]

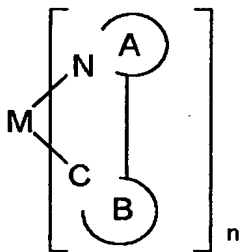
Specification

[Title of the Invention]

Luminescence Device

[Claims]

1. A luminescence device, comprising: an organic compound layer comprising a metal coordination compound represented by the following formula (1):



{wherein M denotes Ir, Rh or Pd; n is 2 or 3; A and B are a nitrogen atom and a carbon atom, respectively, connected to a metal atom M; A and B are cyclic groups containing a nitrogen atom and a carbon atom, respectively, and capable of having a substituent, A and B being connected to each other via a covalent bond, [said substituent being a halogen atom; nitro group; a trialkylsilyl group (of which the alkyl group is a linear or branched alkyl group having 1 - 8 carbon atoms); or a linear or branched alkyl group having 1 - 20 carbon atoms (of which the alkyl group includes one or at least two non-neighboring methylene

groups which can be replaced with -O-, -S-, -CO-, -CO-O-, -O-CO-, -CH=CH- or -C≡C- and includes a hydrogen atom which can be replaced with a fluorine atom] with the proviso that a sum of nitrogen atom and sulfur atom present in ring structures of A and B is at least 2}.

2. A device according to Claim 1, further comprising two electrodes oppositely disposed to sandwich the organic compound layer to constitute an electroluminescence device, wherein a voltage is applied between the pair of electrodes to produce luminescence.

[Detailed Description of the Invention]

[0001]

[Technical Field to which the Invention Pertains]

The present invention relates to a luminescence device using the compound. More specifically, the present invention relates to a luminescence device employing a metal coordination compound as a luminescence material so as to allow stable and high luminescence efficiency.

[0002]

[Prior Art]

An applied research of an organic electroluminescence (EL) device has been

energistically studied as a luminescence device with a high responsiveness and high efficiency.

Basic structures thereof are shown in Figures 1(a) and 1(b) (e.g., as described in "Macromol. Symp.", 125, pp. 1 - 48 (1997)).

[0003]

As shown in Figure 1, the organic EL device generally has a structure including a transparent electrode 14, a metal electrode 11, and a plurality of organic film layers disposed on a transparent substrate 15.

[0004]

Referring to Figure 1(a), the organic layers include a luminescence layer 12 and a hole transport layer 13. The transparent electrode 14 may be formed of a film of ITO (indium tin oxide) having a larger work function to ensure a good hole injection performance from the transparent electrode 14 into the hole transport layer 13. On the other hand, the metal electrode 11 may be formed of a layer of aluminum, magnesium, alloys thereof, etc., having a smaller work function to ensure a good electron injection performance into the organic layer(s). These electrodes may be formed in a thickness of 50 - 200 nm.

[0005]

The luminescence layer 12 may be formed of,

e.g., aluminum quinolinol complex (representative example thereof may include Alq3 shown below having an electron transporting characteristic and a luminescent characteristic. The hole transport layer 13 may be formed of, e.g., triphenyldiamine derivative (representative example thereof may include  $\alpha$ -NPD shown below having an electron donating characteristic.

[0006]

The above-structured device exhibits a rectification characteristic, so that when an electric field is applied between the metal electrode 11 as a cathode and the transparent electrode 14 as an anode, electrons are injected from the metal electrode 11 into the luminescence layer 12 and holes are injected from the transparent electrodes 14.

[0007]

The injected holes and electrons are recombined within the luminescence layer 12 to form excitons, thus producing luminescence. At this time, the hole transport layer 13 functions as an electron-blocking layer to increase a recombination efficiency at the boundary between the luminescence layer 12 and the hole transport layer 13, thus enhancing a luminescence efficiency.

[0008]

Referring to Figure 1(b), an electron

transport layer 16 is disposed between the metal electrode 11 and the luminescence layer 12, whereby an effective carrier blocking performance can be ensured by separating functions of luminescence, electron transport and hole transport, thus allowing efficient luminescence. The electron transport layer 16 may be formed of, e.g., oxadiazole derivatives.

[0009]

In ordinary organic EL devices, heretofore, fluorescence produced during a transition of luminescent center molecule from a singlet excited state to a ground state is used as luminescence. On the other hand, not the above fluorescence (luminescence) via singlet exciton, phosphorescence (luminescence) via triplet exciton has been studied for use in organic EL device. Representative published literature may include Article 1: "Improved energy transfer in electrophosphorescent device" (D.F. O'Brien et al., Applied Physics Letters, Vol. 74, No. 3, pp. 442 - (1999)) and Article 2: "Very high-efficiency green organic light-emitting devices based on electrophosphorescence" (M.A. Baldo et al., Applied Physics Letters, Vol. 75, No. 1, pp. 4 - (1999)).

The EL devices shown in these documents may generally have a four-layer structure shown in Figure 1(c), which structure includes a hole transfer layer 13, a luminescence layer 12, an exciton diffusion-

prevention layer 17, and an electron transport layer 16. Materials used therein are carrier transfer materials and phosphorescent materials shown below. Abbreviations of the respective materials are shown below.

Alq3: aluminum-quinolinol complex,

$\alpha$ -NPD: N4,N4'-di-naphthalene-1-yl-N4,N4'-diphenyl-biphenyl-4,4'-diamine,

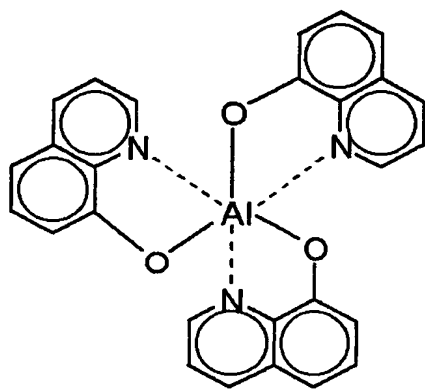
CBP: 4,4'-N,N'-dicarbazole-biphenyl,

BCP: 2,9-dimethyl-4,7-diphenyl-1,10-phenanthroline,

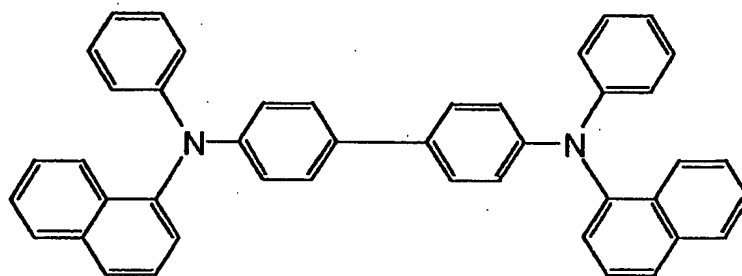
PtEOP: platinum-octaethyl porphine complex, and

Ir(ppy)<sub>3</sub>: iridium-phenylpyridine complex.

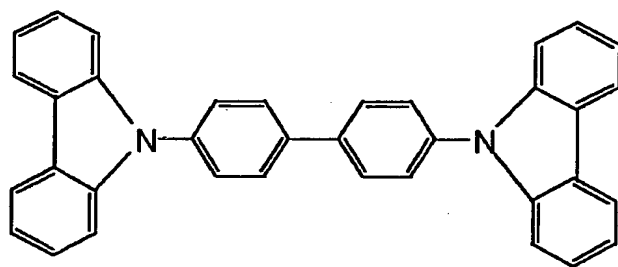
[0011]



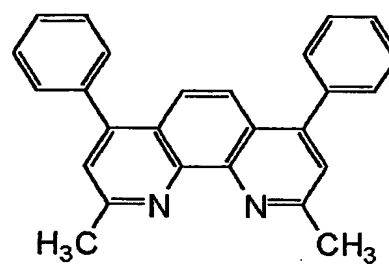
Alq3



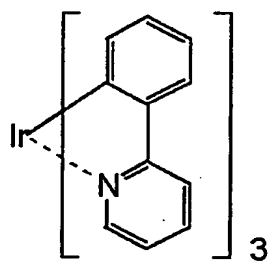
$\alpha$ -NPD



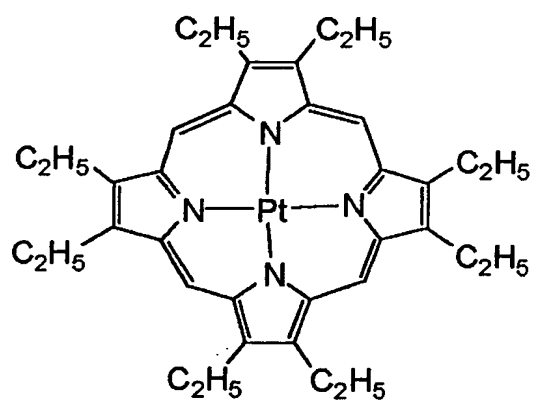
CBP



BCP



Ir(ppy)<sub>3</sub>



PtOEP

[0012]

In both the Articles 1 and 2, higher efficiencies have been achieved by using a layer structure including a hole transport layer 13 of  $\alpha$ -NPD, an electron transport layer 16 of Alq3, an exciton diffusion-prevention layer 17 of BPC, and a luminescence layer 12 of a mixture of CPB as a host material with Ir(ppy)<sub>3</sub> or PtOEP as a (guest) phosphorescence material doped into CBP at a concentration of ca. 6 wt. %.

[0013]

The reason why the phosphorescence (luminescence) material has particularly attracted notice is that the phosphorescence material is expected to provide a higher luminescence efficiency in principle. This is because excitons produced by recombination of carriers comprise singlet excitons and triplet excitons presented in a ratio of 1:3. In the conventional organic EL device, fluorescence at the time of transition from the singlet excited state to the ground state is utilized as luminescence. A resultant luminescence efficiency is 25 % (which is an upper limit) based on the number of the produced excitons in principle. However, if phosphorescence from excitons generated from the triplet excited state is used, a resultant luminescence efficiency is expected to be at least three times that of the case

of fluorescence in principle. In addition thereto, if intersystem crossing from the singlet excited state which is a higher energy level to the triplet excited state is taken into consideration, the luminescence efficiency of phosphorescence can be expected to be 100 % (four times that of fluorescence) in principle.

[0014]

Other documents describing phosphorescence from the triplet excited state may include, e.g., Japanese Laid-Open Patent Application (JP-A) 11-329739 (organic EL device and production process thereof), JP-A 11-256148 (luminescent device and organic EL device) and JP-A 8-319482 (organic electroluminescent device).

[0015]

[Problems to be Solved by the Invention]

In the above-mentioned organic EL device utilizing phosphorescence have accompanied with a problem of luminescent deterioration particularly in an energized state. The reason for luminescent deterioration has not been clarified as yet but may be attributable to such a phenomenon that the life of triplet exciton is generally longer than that of singlet exciton by at least three digits, so that molecule is placed in a higher-energy state for a long period to cause reaction with ambient substance, formation of exciplex or excimer, change in minute

molecular structure, structural change of ambient substance, etc.

[0016]

Anyway, the (electro)phosphorescence device is expected to provide a higher luminescence efficiency but on the other hand, the device is problematic in terms of the (luminescent) deterioration in energized state.

[0017]

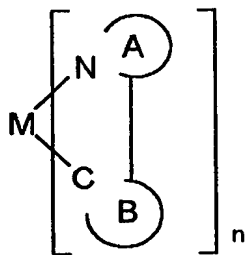
Accordingly, an object of the present invention is to provide a luminescence device capable of providing a high-efficiency luminescent, keeping a high brightness (or luminance) for a long period, and less deterioration (in luminescence) in energized state.

[0018]

[Means for Solving the Problems]

That is, the present invention provides a luminescence device characterized in that an organic compound layer comprising a metal coordination compound represented by the following formula (1):

[0019]



[0020]

{wherein M denotes Ir, Rh or Pd; n is 2 or 3; A and B are a nitrogen atom and a carbon atom, respectively, connected to a metal atom M; A and B are cyclic groups containing a nitrogen atom and a carbon atom, respectively, and capable of having a substituent, A and B being connected to each other via a covalent bond, [said substituent being a halogen atom; nitro group; a trialkylsilyl group (of which the alkyl group is a linear or branched alkyl group having 1 - 8 carbon atoms); or a linear or branched alkyl group having 1 - 20 carbon atoms (of which the alkyl group includes one or at least two non-neighboring methylene groups which can be replaced with -O-, -S-, -CO-, -CO-O-, -O-CO-, -CH=CH- or -C≡C- and includes a hydrogen atom which can be replaced with a fluorine atom] with the proviso that a sum of nitrogen atom and sulfur atom present in ring structures of A and B is at least 2}.

[0021]

The luminescence device described above, may preferably include two electrodes oppositely disposed to sandwich the organic compound layer to constitute an electroluminescence device, wherein a voltage is applied between the pair of electrodes to produce luminescence.

[0022]

[Embodiments for Practicing the Invention]

In the case where a luminescence layer is formed of a carrier transporting host material and a phosphorescent guest material, a process of producing phosphorescence may generally involve the following steps:

- (1) transport of electron and hole within a luminescence layer,
- (2) formation of exciton of the host material,
- (3) transmission of excited energy between host material molecules,
- (4) transmission of excited energy from the host material molecule to the guest material molecule,
- (5) formation of triplet exciton of the guest material, and
- (6) production of phosphorescence during transition from the triplet excited state to the ground state of the guest material.

[0023]

In the respective steps, desired energy transmission and luminescence may generally be caused based on various quenching and competition.

[0024]

In order to improve a luminescence efficiency of the EL device, it is needless to say that a luminescence center material per se provides a higher yield of luminescence quantum. However, an efficient

energy transfer between host material molecules or between host material molecule and guest material molecule is also an important factor. Further, the reason for the above-described luminescent deterioration in energized state is not clarified as yet but may presumably relate to the luminescent center material per se or an environmental change thereof by its ambient molecular structure.

[0025]

For this reason, the present inventors have extensively investigated an effect of use of the metal coordination compound of formula (1) as the luminescent center material and have found that the compound produces a high-efficiency luminescence and retains a high brightness (luminance) for a long period.

[0026]

The metal coordination compound used in the present invention produces phosphorescence (luminescence) and is assumed to have a lowest excited state which is an MLCT\* (metal-to-ligand charge transfer) state as a triplet (excited state). The phosphorescence is produced during the transition from these states to the ground state.

[0027]

The luminescent material of the present invention has been found to provide a higher

phosphorescence yield of 0.15 - 0.9 and a shorter phosphorescence life of 1 - 30  $\mu$ sec.

The shorter phosphorescence life is a condition for providing a resultant EL device with a higher luminescence efficiency. More specifically, if the phosphorescence life is longer, molecules placed in their triplet excited state which is a waiting state for phosphorescence are increased to result in a problem of lowering the resultant luminescence efficiency particularly at a higher current density. The material of the present invention is a suitable luminescent material for an EL device with a higher phosphorescence yield and a shorter phosphorescence life. Further, due to realization of the shorter phosphorescence life leading to a shorter time period wherein the molecules stay in the triplet excited state, i.e. a higher energy state, so that the resultant EL device with improved durability and less deterioration in device characteristic.

[0028]

Actually, even in energization test, when the luminescent material of the present invention was used, it exhibited high stability.

[0029]

Further, in the ligand, according to the present invention, a plurality of nitrogen atom(s) and/or sulfur atom(s) in total in two ring structures,

so that it becomes possible to decrease an energy gap, thus allowing a long-(emission) wavelength luminescence. This may be understood as follows.

In the case where HOMO (highest occupied molecular orbital) and LUMO (lowest unoccupied molecular orbital) of electron orbital for the metal and those (HOMO and LUMO) of electron orbital for the ligands are separately considered for convenience, energies of HOMO and LUMO for the ligands are lowered by the presence of nitrogen atom(s) and/or sulfur atom(s) to decrease an energy gap between HOMO for the metal and LUMO for the ligands, thus allowing luminescence from the MLCT\* state being the lowest excited state at a longer (emission) wavelength. Accordingly, the metal coordination compound of the present invention is suitable luminescent material for luminescence at longer wavelength (orange to red).

[0030]

Further, the luminescent material of the present invention contains a plurality of nitrogen atom(s) and/or sulfur atom(s) in the ligands, so that electron clouds may presumably be considerably protruded from the ligands to promote interaction with other molecules via these atoms, thus improving energy transfer efficiency. As a result, it becomes possible to realize a luminescent material, for longer wavelength (orange to red) with high efficiency and

stability, which has not been conventionally provided.

[0031]

In the case of phosphorescent (luminescent) material, luminescent characteristics are largely depend on its molecular environment. In the case of the fluorescent material, essential properties of the luminescent material are studied based on photoluminescence. For this reason, results of photoluminescence of the phosphorescent material do not reflect luminescent characteristics of the resultant EL device in many cases since the luminescent characteristics in the case of the phosphorescent material depend on a magnitude of polarity of ambient host material molecules, ambient temperature, state of solid or liquid, etc. Accordingly, it is generally difficult to expect the resultant EL characteristics (for the phosphorescent material) by simply removing a part of characteristics from photoluminescence results.

[0032]

Further, as shown in Examples appearing hereinafter, the compound of the present invention has been clarified to exhibit excellent performance even in terms of stability in an energization durability test.

[0033]

As shown in Figure 1, the luminescence device of the present invention may preferably be an

electroluminescent device wherein an organic layer comprising a metal coordination compound is sandwiched between oppositely disposed two electrodes between which are supplied with a voltage to produce luminescence.

[0034]

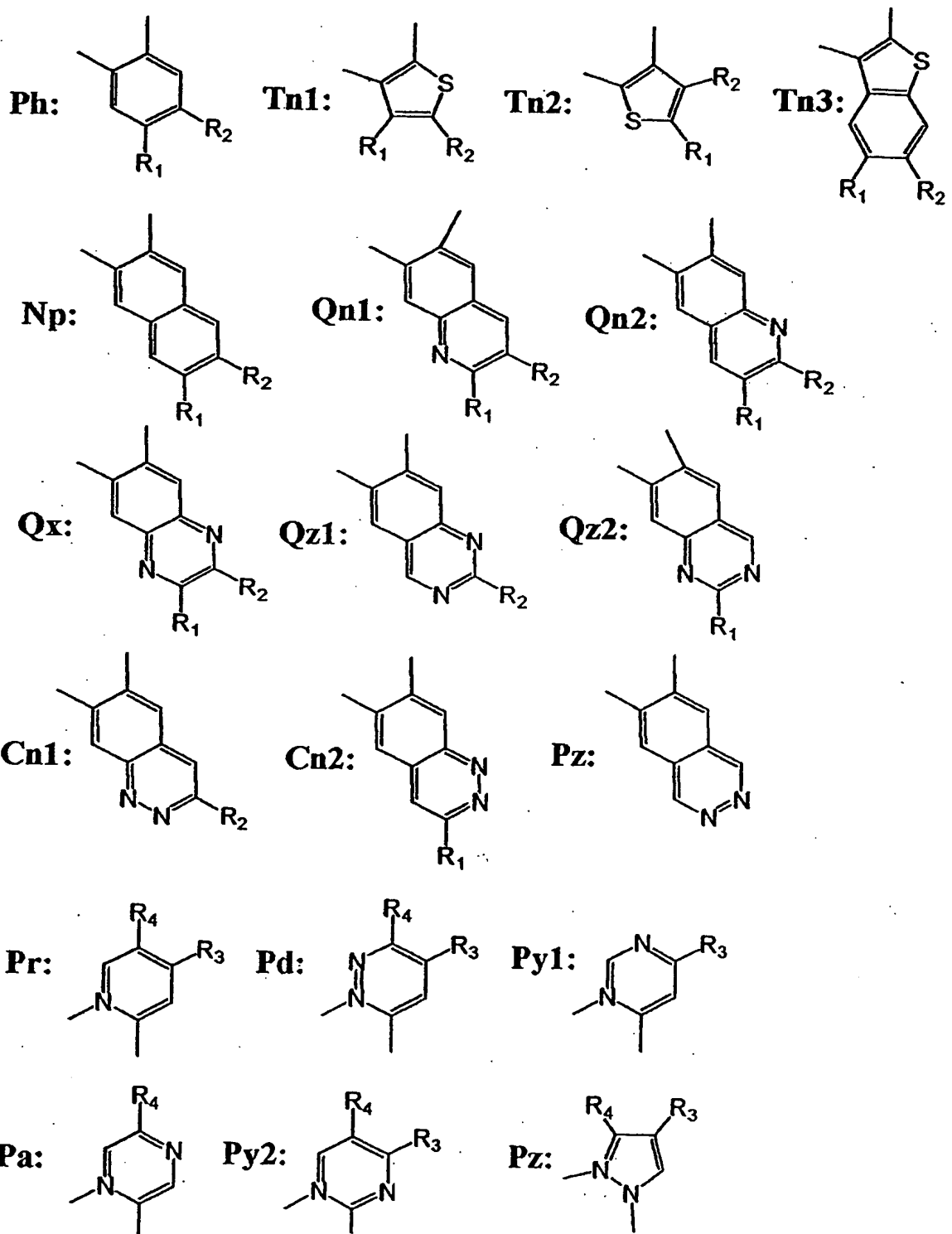
The high-efficiency luminescence device according to the present invention may be applicable to devices required to allow energy saving and high luminance. Applied examples thereof may include display apparatus and illumination apparatus, a light source for printers, and backlight (unit) for a liquid crystal display apparatus. As the display apparatus, it is possible to provide a flat panel display apparatus capable of exhibiting an excellent energy saving performance, a high visibility and a good lightweight property. With respect to the light source, it becomes possible to replace a laser light source of laser beam printer currently used widely with the luminescence device according to the present invention. An independently addressable device is arranged in an array and desired exposure of light to a photosensitive drum is effected to form an image. By using the device of the present invention, it becomes possible to considerably reducing the volume (size) of image forming apparatus. With respect to the illumination apparatus and backlight (unit), an energy

saving effect by the present invention can be expected.

[0035]


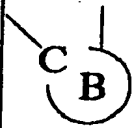
Hereinbelow, specific structural formulas of the metal coordination compound used in the present invention will be shown in Tables 1 - 9. However, these merely show representative examples and the present invention is not limited thereto. Ph to Pz used in Tables 1 - 9 represent structures shown below.

[0036]




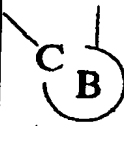
[0037]

[Table 1]

No	M	n			R1	R2	R3	R4
1	Ir	3	Pr	Tn1	H	H	H	H
2	Ir	3	Pr	Tn2	H	H	H	H
3	Ir	3	Pr	Tn3	H	H	H	H
4	Ir	3	Pr	Qn1	H	H	H	H
5	Ir	3	Pr	Qn2	H	H	H	H
6	Ir	3	Pr	Qx	H	H	H	H
7	Ir	3	Pr	Qz1	H	H	H	H
8	Ir	3	Pr	Qz2	H	H	H	H
9	Ir	3	Pr	Cn1	H	H	H	H
10	Ir	3	Pr	Cn2	H	H	H	H
11	Ir	3	Pr	Pz	H	H	H	H
12	Ir	3	Pd	Ph	H	H	H	H
13	Ir	3	Pd	Tn1	H	H	H	H
14	Ir	3	Pd	Tn2	H	H	H	H
15	Ir	3	Pd	Tn3	H	H	H	H
16	Ir	3	Pd	Np	H	H	H	H
17	Ir	3	Pd	Qn1	H	H	H	H
18	Ir	3	Pd	Qn2	H	H	H	H
19	Ir	3	Pd	Qx	H	H	H	H
20	Ir	3	Pd	Qz1	H	H	H	H
21	Ir	3	Pd	Qz2	H	H	H	H
22	Ir	3	Pd	Cn1	H	H	H	H
23	Ir	3	Pd	Cn2	H	H	H	H
24	Ir	3	Pd	Pz	H	H	H	H
25	Ir	3	Py1	Ph	H	H	H	H


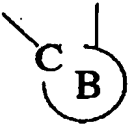
[0038]

[Table 2]

No	M	n			R1	R2	R3	R4
26	Ir	3	Py1	Tn1	H	H	H	H
27	Ir	3	Py1	Tn2	H	H	H	H
28	Ir	3	Py1	Tn3	H	H	H	H
29	Ir	3	Py1	Np	H	H	H	H
30	Ir	3	Py1	Qn1	H	H	H	H
31	Ir	3	Py1	Qn2	H	H	H	H
32	Ir	3	Pa	Ph	H	H	H	H
33	Ir	3	Pa	Tn1	H	H	H	H
34	Ir	3	Pa	Tn2	H	H	H	H
35	Ir	3	Pa	Np	H	H	H	H
36	Ir	3	Pa	Qn1	H	H	H	H
37	Ir	3	Pa	Qn2	H	H	H	H
38	Ir	3	Py2	Ph	H	H	H	H
39	Ir	3	Py2	Tn1	H	H	H	H
40	Ir	3	Py2	Tn2	H	H	H	H
41	Ir	3	Py2	Tn3	H	H	H	H
42	Ir	3	Py2	Np	H	H	H	H
43	Ir	3	Py2	Qn1	H	H	H	H
44	Ir	3	Py2	Qn2	H	H	H	H
45	Ir	3	Pz	Ph	H	H	H	H
46	Ir	3	Pz	Tn1	H	H	H	H
47	Ir	3	Pz	Tn2	H	H	H	H
48	Ir	3	Pz	Np	H	H	H	H
49	Ir	3	Pz	Qn1	H	H	H	H
50	Ir	3	Pz	Qn2	H	H	H	H


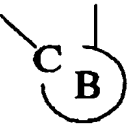
[0039]

[Table 3]

No	M	n			R1	R2	R3	R4
51	Rh	3	Pr	Tn1	H	H	H	H
52	Rh	3	Pr	Tn2	H	H	H	H
53	Rh	3	Pr	Tn3	H	H	H	H
54	Rh	3	Pr	Qn1	H	H	H	H
55	Rh	3	Pr	Qn2	H	H	H	H
56	Rh	3	Pr	Qx	H	H	H	H
57	Rh	3	Pr	Qz1	H	H	H	H
58	Rh	3	Pr	Qz2	H	H	H	H
59	Rh	3	Pr	Cn1	H	H	H	H
60	Rh	3	Pr	Cn2	H	H	H	H
61	Rh	3	Pr	Pz	H	H	H	H
62	Rh	3	Pd	Ph	H	H	H	H
63	Rh	3	Pd	Tn1	H	H	H	H
64	Rh	3	Pd	Tn2	H	H	H	H
65	Rh	3	Pd	Tn3	H	H	H	H
66	Rh	3	Pd	Np	H	H	H	H
67	Rh	3	Pd	Qn1	H	H	H	H
68	Rh	3	Pd	Qn2	H	H	H	H
69	Rh	3	Pd	Qx	H	H	H	H
70	Rh	3	Pd	Qz1	H	H	H	H
71	Rh	3	Pd	Qz2	H	H	H	H
72	Rh	3	Pd	Cn1	H	H	H	H
73	Rh	3	Pd	Cn2	H	H	H	H
74	Rh	3	Pd	Pz	H	H	H	H
75	Rh	3	Py1	Ph	H	H	H	H


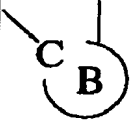
[0040]

[Table 4]

No	M	n			R1	R2	R3	R4
76	Rh	3	Py1	Tn1	H	H	H	H
77	Rh	3	Py1	Tn2	H	H	H	H
78	Rh	3	Py1	Tn3	H	H	H	H
79	Rh	3	Py1	Np	H	H	H	H
80	Rh	3	Py1	Qn1	H	H	H	H
81	Rh	3	Py1	Qn2	H	H	H	H
82	Rh	3	Pa	Ph	H	H	H	H
83	Rh	3	Pa	Tn1	H	H	H	H
84	Rh	3	Pa	Tn2	H	H	H	H
85	Rh	3	Pa	Np	H	H	H	H
86	Rh	3	Pa	Qn1	H	H	H	H
87	Rh	3	Pa	Qn2	H	H	H	H
88	Rh	3	Py2	Ph	H	H	H	H
89	Rh	3	Py2	Tn1	H	H	H	H
90	Rh	3	Py2	Tn2	H	H	H	H
91	Rh	3	Py2	Tn3	H	H	H	H
92	Rh	3	Py2	Np	H	H	H	H
93	Rh	3	Py2	Qn1	H	H	H	H
94	Rh	3	Py2	Qn2	H	H	H	H
95	Rh	3	Pz	Ph	H	H	H	H
96	Rh	3	Pz	Tn1	H	H	H	H
97	Rh	3	Pz	Tn2	H	H	H	H
98	Rh	3	Pz	Np	H	H	H	H
99	Rh	3	Pz	Qn1	H	H	H	H
100	Rh	3	Pz	Qn2	H	H	H	H

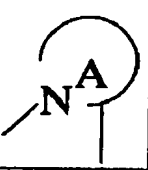
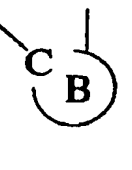
[0041]

[Table 5]

No	M	n			R1	R2	R3	R4
101	Pd	2	Pr	Tn1	H	H	H	H
102	Pd	2	Pr	Tn2	H	H	H	H
103	Pd	2	Pr	Tn3	H	H	H	H
104	Pd	2	Pr	Qn1	H	H	H	H
105	Pd	2	Pr	Qn2	H	H	H	H
106	Pd	2	Pr	Qx	H	H	H	H
107	Pd	2	Pr	Qz1	H	H	H	H
108	Pd	2	Pr	Qz2	H	H	H	H
109	Pd	2	Pr	Cn1	H	H	H	H
110	Pd	2	Pr	Cn2	H	H	H	H
111	Pd	2	Pr	Pz	H	H	H	H
112	Pd	2	Pd	Ph	H	H	H	H
113	Pd	2	Pd	Tn1	H	H	H	H
114	Pd	2	Pd	Tn2	H	H	H	H
115	Pd	2	Pd	Tn3	H	H	H	H
116	Pd	2	Pd	Np	H	H	H	H
117	Pd	2	Pd	Qn1	H	H	H	H
118	Pd	2	Pd	Qn2	H	H	H	H
119	Pd	2	Pd	Qx	H	H	H	H
120	Pd	2	Pd	Qz1	H	H	H	H
121	Pd	2	Pd	Qz2	H	H	H	H
122	Pd	2	Pd	Cn1	H	H	H	H
123	Pd	2	Pd	Cn2	H	H	H	H
124	Pd	2	Pd	Pz	H	H	H	H
125	Pd	2	Py1	Ph	H	H	H	H


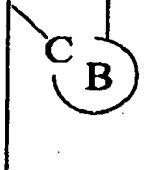
[0042]

[Table 6]

No	M	n			R1	R2	R3	R4
126	Pd	2	Py1	Tn1	H	H	H	H
127	Pd	2	Py1	Tn2	H	H	H	H
128	Pd	2	Py1	Tn3	H	H	H	H
129	Pd	2	Py1	Np	H	H	H	H
130	Pd	2	Py1	Qn1	H	H	H	H
131	Pd	2	Py1	Qn2	H	H	H	H
132	Pd	2	Pa	Ph	H	H	H	H
133	Pd	2	Pa	Tn1	H	H	H	H
134	Pd	2	Pa	Tn2	H	H	H	H
135	Pd	2	Pa	Np	H	H	H	H
136	Pd	2	Pa	Qn1	H	H	H	H
137	Pd	2	Pa	Qn2	H	H	H	H
138	Pd	2	Py2	Ph	H	H	H	H
139	Pd	2	Py2	Tn1	H	H	H	H
140	Pd	2	Py2	Tn2	H	H	H	H
141	Pd	2	Py2	Tn3	H	H	H	H
142	Pd	2	Py2	Np	H	H	H	H
143	Pd	2	Py2	Qn1	H	H	H	H
144	Pd	2	Py2	Qn2	H	H	H	H
145	Pd	2	Pz	Ph	H	H	H	H
146	Pd	2	Pz	Tn1	H	H	H	H
147	Pd	2	Pz	Tn2	H	H	H	H
148	Pd	2	Pz	Np	H	H	H	H
149	Pd	2	Pz	Qn1	H	H	H	H
150	Pd	2	Pz	Qn2	H	H	H	H

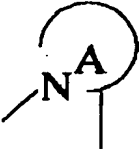
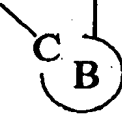
[0043]

[Table 7]

No	M	n			R1	R2	R3	R4
151	Ir	3	Pr	Tn1	H	Si(CH <sub>3</sub> ) <sub>3</sub>	H	H
152	Ir	3	Pr	Tn1	H	SiC <sub>2</sub> H <sub>5</sub>	H	H
153	Ir	3	Pr	Tn1	H	OCH <sub>3</sub>	H	H
154	Ir	3	Pr	Tn1	H	H	H	F
155	Ir	3	Pr	Tn1	H	H	F	F
156	Ir	3	Pr	Tn1	H	H	H	C <sub>8</sub> H <sub>11</sub>
157	Ir	3	Pr	Tn1	H	H	H	OCH <sub>3</sub>
158	Ir	3	Pr	Tn1	H	H	OC <sub>4</sub> H <sub>9</sub>	H
159	Ir	3	Pr	Tn1	H	H	H	OCF <sub>3</sub>
160	Ir	3	Pr	Tn1	H	H	H	SC <sub>2</sub> H <sub>5</sub>
161	Ir	3	Pr	Tn1	H	H	H	COOC <sub>2</sub> H <sub>5</sub>
162	Ir	3	Pr	Tn1	H	H	H	COOC <sub>2</sub> H <sub>5</sub>
163	Ir	3	Pr	Tn1	H	H	H	OCOC <sub>2</sub> H <sub>5</sub>
164	Ir	3	Py1	Np	H	F	H	H
165	Ir	3	Pa	Np	H	OCH <sub>3</sub>	H	H
166	Ir	3	Pz	Ph	H	H	H	CH <sub>3</sub>
167	Ir	3	Pd	Ph	H	NO <sub>2</sub>	H	H
168	Ir	3	Py1	Ph	H	NO <sub>2</sub>	H	H
169	Ir	3	Pa	Ph	H	NO <sub>2</sub>	H	H
170	Ir	3	Py2	Ph	H	F	H	H
171	Ir	3	Pz	Ph	H	F	H	H
172	Ir	3	Pz	Ph	H	CH <sub>2</sub> CH=CH CH <sub>2</sub> CH <sub>2</sub>	H	H
173	Ir	3	Pz	Ph	H	CH <sub>2</sub> C≡ CCH <sub>2</sub> CH <sub>3</sub>	H	H
174	Rh	3	Pr	Tn1	H	Si(CH <sub>3</sub> ) <sub>3</sub>	H	H
175	Rh	3	Pr	Tn1	H	OCH <sub>3</sub>	H	H

[0044]

[Table 8]

No	M	n			R1	R2	R3	R4
176	Rh	3	Pr	Tn1	H	H	H	F
177	Rh	3	Pr	Tn1	H	H	F	F
178	Rh	3	Pr	Tn1	H	H	H	C <sub>6</sub> H <sub>11</sub>
179	Rh	3	Pr	Tn1	H	H	H	OCH <sub>3</sub>
180	Rh	3	Pr	Tn1	H	H	OC <sub>4</sub> H <sub>9</sub>	H
181	Rh	3	Pr	Tn1	H	H	H	OCF <sub>3</sub>
182	Rh	3	Pr	Tn1	H	H	H	SC <sub>2</sub> H <sub>5</sub>
183	Rh	3	Pr	Tn1	H	H	H	COC <sub>2</sub> H <sub>5</sub>
184	Rh	3	Py1	Np	H	F	H	H
185	Rh	3	Pa	Np	H	OCH <sub>3</sub>	H	H
186	Rh	3	Pz	Ph	H	H	H	CH <sub>3</sub>
187	Rh	3	Pz	Tn1	H	F	H	H
188	Pd	2	Pr	Tn1	H	Si(CH <sub>3</sub> ) <sub>3</sub>	H	H
189	Pd	2	Pr	Tn1	H	OCH <sub>3</sub>	H	H
190	Pd	2	Pr	Tn1	H	H	H	F
191	Pd	2	Pr	Tn1	H	H	F	F
192	Pd	2	Pr	Tn1	H	H	H	C <sub>6</sub> H <sub>11</sub>
193	Pd	2	Pr	Tn1	H	H	H	OCH <sub>3</sub>
194	Pd	2	Pr	Tn1	H	H	OC <sub>4</sub> H <sub>9</sub>	H
195	Pd	2	Pr	Tn1	H	H	H	OCF <sub>3</sub>
196	Pd	2	Pr	Tn1	H	H	H	SC <sub>2</sub> H <sub>5</sub>
197	Pd	2	Pr	Tn1	H	H	H	COC <sub>2</sub> H <sub>5</sub>
198	Pd	2	Py1	Np	H	F	H	H
199	Pd	2	Pa	Np	H	OCH <sub>3</sub>	H	H
200	Pd	2	Pz	Ph	H	H	H	CH <sub>3</sub>
201	Pd	2	Pz	Tn1	H	F	H	H

[0045]

[Examples]

(Examples 1 - 10 and Comparative Example 1)

A common portion of device preparation steps employed in the present invention will be described.

[0046]

As a device structure, a device shown in Figure 1(b) having three organic layers was used. On a glass substrate (transparent substrate 15), a 100 nm-thick film (transparent electrode 14) of ITO (indium tin oxide) was formed, followed by patterning to have an opposing electrode area of 3 mm<sup>2</sup>.

On the ITO-formed substrate, organic layers and metal electrode layers shown below were successively formed by vacuum (vapor) deposition using resistance heating in a vacuum chamber (10<sup>-4</sup> Pa).

Organic layer 1 (hole transport layer 13) (40 nm):  $\alpha$ -NPD

Organic layer 2 (luminescence layer 12) (30 nm): CBP:predetermined coordination compound (weight proportion: 5 wt. %)

Organic layer 3 (electron transport layer 16) (30 nm): Alq3

Metal electrode layer 1 (15 nm): Al-Li alloy (Li content = 1.8 wt. %)

Metal electrode layer 2 (100 nm): Al

[0047]

An electric field (voltage) was applied, so as to provide each device with the same current value, to each luminescence device having the ITO electrode (as an anode) and the Al electrode (as a cathode), to measure a change in luminance (brightness) with time. The current amount was set to  $70 \text{ mA/cm}^2$  and the respective devices showed luminances in the range of  $80 - 240 \text{ cd/m}^2$  at an initial stage required for decreasing.

[0048]

For measurement, each luminescence device was taken out of the vacuum chamber and was subjected to the measurement in an atmosphere of dry nitrogen gas stream so as to remove device deterioration factors, such as oxygen and moisture (water content).

[0049]

The results of continuous energization test of devices using the respective compound are shown in Table 10. As a comparative example, an experiment of the device using  $\text{Ir(ppy)}_3$  as the luminescence material was conducted. The device (using  $\text{Ir(ppy)}_3$ ) clearly provided shorter luminance half-life than the devices using the luminescent materials of the present invention, so that it becomes possible to provide a device having a high durability based on a stability of the materials of the present invention.

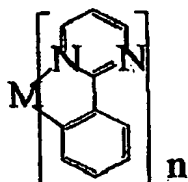
[0050]

[Table 10]

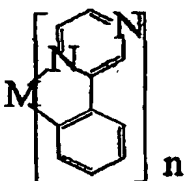
Ex. and Comp.Ex. No.	Luminescence material			Luminescence half-life (hours)
	Chem. formula No.	M (metal atom)	n	
Ex. 1	21	Ir	3	550
Ex. 2	22	Ir	3	900
Ex. 3	23	Ir	3	600
Ex. 4	24	Ir	3	650
Ex. 5	25	Ir	3	950
Ex. 6	26	Ir	3	800
Ex. 7	27	Ir	3	850
Ex. 8	28	Ir	3	600
Ex. 9	29	Ir	3	750
Ex. 10	30	Ir	3	900
Comp. Ex. 1		Ir(ppy) <sub>3</sub>		350

[0051]

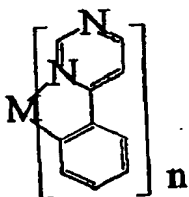
Chem. formula 21



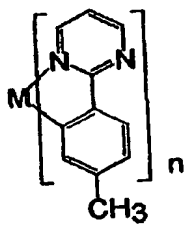
Chem. formula 22



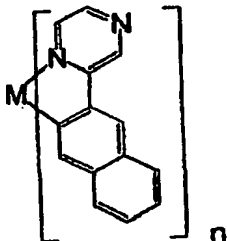
Chem. formula 23



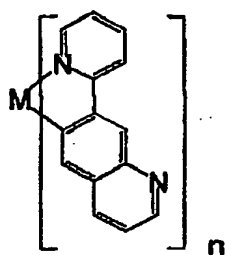
Chem. formula 24



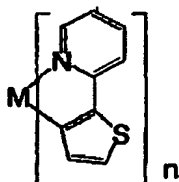
Chem. formula 25



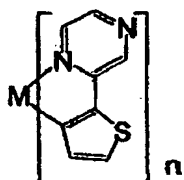
Chem. formula 26



Chem. formula 27



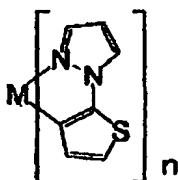
Chem. formula 28



Chem. formula 29



Chem. formula 30



[0052]

[Effect of the Invention]

As described hereinabove, the metal coordination compound used in the present invention has a higher phosphorescence luminescence efficiency and a shorter phosphorescence life, thus being suitable for longer-waveform luminescence (orange to red) and being suitable as a luminescence material for an EL device.

[0053]

Further, the luminescence device using the metal coordination compound according to the present invention is an excellent device which produces a high-efficiency luminescence, retains a high luminescence for a long period of time, and provides less deterioration in energized state.

[Brief Description of the Drawings]

Figure 1 is a schematic sectional view of a layer structure of a luminescence device of the present invention.

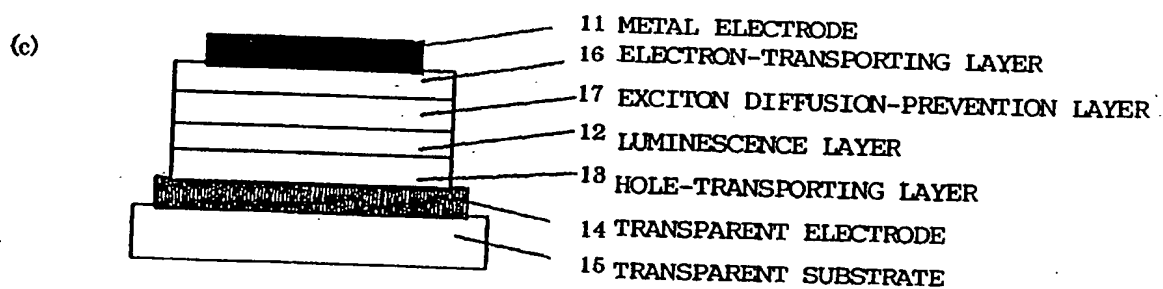
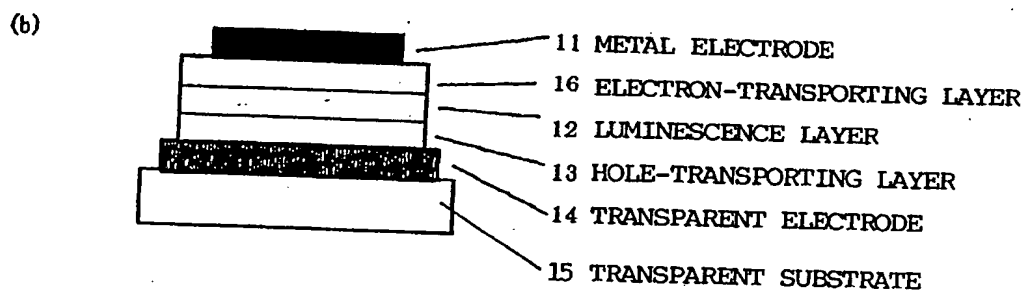
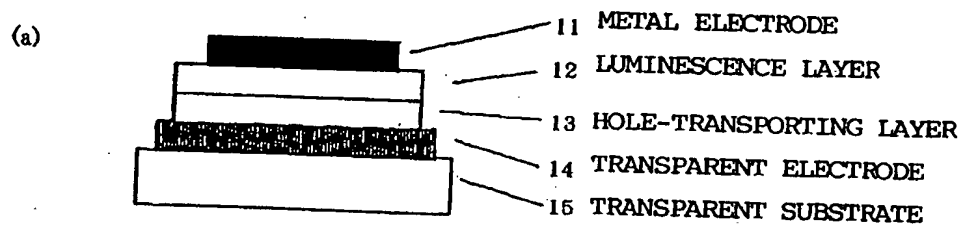
[Explanation of Symbols]

- 11: metal electrode
- 12: luminescence layer
- 13: hole transport layer
- 14: transparent electrode
- 15: transparent substrate

16: electron transport layer

17: exciton diffusion prevention layer

FIG. 1



[Document]

Abstract

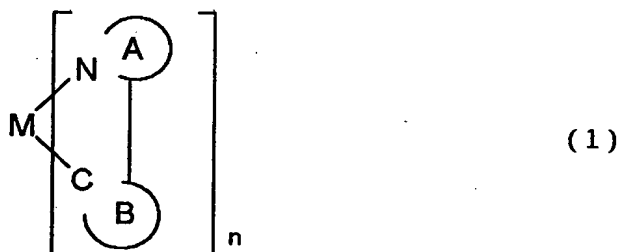
[Abstract]

[Problem]

To provide a luminescence device which produces a high-efficiency luminescence, maintains a high luminance for a long period of time, and provides less deterioration in energized state.

[Solution Means]

A luminescence device characterized by including an organic compound layer containing a metal coordination compound represented by the following formula (1):



{wherein M denotes Ir, Rh or Pd; n is 2 or 3; A and B are a nitrogen atom and a carbon atom, respectively, connected to a metal atom M; A and B are cyclic groups containing a nitrogen atom and a carbon atom, respectively, and capable of having a substituent, A and B being connected to each other via a covalent bond, [said substituent being a halogen atom; nitro group; a trialkylsilyl group (of which the alkyl group is a linear or branched alkyl group having 1 - 8

carbon atoms); or a linear or branched alkyl group having 1 - 20 carbon atoms (of which the alkyl group includes one or at least two non-neighboring methylene groups which can be replaced with -O-, -S-, -CO-, -CO-O-, -O-CO-, -CH=CH- or -C≡C- and includes a hydrogen atom which can be replaced with a fluorine atom] with the proviso that a sum of nitrogen atom and sulfur atom present in ring structures of A and B is at least 2}.

[Selected Figure]

Figure 1